

Hydrogeochemical characteristics of interstitial water and overlying water in the lacustrine environment

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Abstract This research was conducted in the lacustrine environment of Lake Wigry, one of the largest and deepest lakes in Poland. The main objective was to determine the relationship between the chemical composition of interstitial water, overlying water and bottom sediment in the different sedimentation conditions. Therefore, seven research positions were selected, taking into account different zones of lake and lithological type of deposits. Overlying water samples taken from zones with carbonate type of sediment (lacustrine chalk or calcareous gyttja) were characterized by slightly alkaline pH, similar hydrogeochemical type ($\text{HCO}_3\text{--Ca--Mg}$, $\text{HCO}_3\text{--SO}_4\text{--Ca--Mg}$), low values of mineralization and low metal concentrations. The situation is different with the water taken from the zone of organic: detritus gyttja or dy deposits. Overlying water collected from the Hańczańska Bay (estuary of Czarna Hańcza River) had the highest mineralization 552 mg/L, $\text{HCO}_3\text{--Ca}$ type and low metal concentrations, while water collected from the dystrophic lake was $\text{SO}_4\text{--Ca--Mg}$ type, low mineralized and acidic type. Studies of interstitial solutions collected from different lithological type of sediments showed noticeable chemical composition differences, both between the carbonate and organic sediments in the same reservoir, as well as in the same kind of sediment but exposed to different external conditions. The highest mineralization

(2032 mg/L) was generally found in the interstitial water obtained from the detritus gyttja localized in the shallow positions, which are more exposed to pollution. The highest trace metals and nutrients content in the sediment and interstitial water found in the Czarna Hańcza River estuary were a result of the input from increasing industrial wastewater, domestic sewage discharge and runoff from agricultural soil. A greater resistance to adsorption of contaminants characterizes sediments composed of the lacustrine chalk; mineralization of their pore solutions was significantly lower. This fact is probably also affected by very low mineralization and chemical composition of overlying water and sediment.

Keywords Lacustrine sediment · Interstitial water · Chemical composition · Lake Wigry · Dystrophic lake

Introduction

Interstitial water constitutes the link between the aquatic sediment and the overlying water, with the sediment–water interface associated with steep gradients in density, particle and solution composition, activities of chemical species, pH, redox potential and biological activity between two phases (Santschi 1990). In the assessment of the aquatic environment, it is important to determine interactions between the direct contact separating sediment and water. For this, it is necessary to recognize the chemical composition of the interstitial water that fills the pore spaces of solid particles within sediments of a body of water.

Knowledge of the chemical composition and its temporal and spatial variation of the interstitial water may explain the origin of water and its palaeohydrogeological processes. This can prove effective in assessing conditions

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of circulation and exchange rate of groundwater as well (Szczepańska et al. 1980). It is also useful in determining sediments' participation in overlying water contamination (Aggett and O'Brien 1985). This is particularly important in the case of reservoir dredging when there is a potential risk of migration of contaminants into the water and/or their mobilization within the sediment (Brannon et al. 1980). Generally, there are two groups of factors that are the main determinants of pore water chemistry. The first factor determines the initial composition of the water at the time of the sediment formation, for example, geographical and hydrochemical environment and conditions in the reservoir. The second are the factors and processes leading to changes in the original composition of water, for example, cation exchange reactions between the interstitial water and exchangeable rocks complex, processes of diffusion, ion movement or participation of the pore water in the minerals formation (Myślińska 1974).

Mineralization and chemical composition of interstitial water depends therefore not only on the palaeogeographic conditions of their formation, but primarily on a number of physical and chemical processes occurring in the sediments from the moment of sedimentation. Among these aforementioned conditions, the most important are (Schulz 2006): substance removing within interstitial water by precipitation or adsorption and substance releasing into interstitial water from the solid phase (i.e. by dissolution or desorption or transporting of dissolved substances by diffusion within interstitial water and across the sediment–water interface). These processes can sometimes completely change the initial composition of interstitial water. Factors that might affect the chemical bioavailability in interstitial solutions include sorption to test container surfaces and reduction through shifts in pH (Adams et al. 2000). Chemical bioavailability can be also reduced by organic carbon dissolved in interstitial water (Suedel et al. 1993; Williamson and Burgess 2000). Maassen et al. (2005) presented that sediment and pore water both reflect the trophic characteristic of the water body and are, in turn, also strongly affected by it. They proposed to include the tested water and sediment into common trophic evaluation systems.

The diagenetic processes in sediment are also the reason for the frequent variation of the chemical characteristics of interstitial and overlying water (Salomons et al. 1987). Interstitial water is a very sensitive indicator of diagenetic reactions occurring within the aquatic sediment and shows marked changes in chemical composition, while changes in the sediments' chemical characteristics may be unnoticeable (Berner 1980). Temporal changes in the interstitial water may also depend on seasonal changes in temperature, sedimentation rate and the biomass production (Song and Müller 1995).

The main goal of this research was to assess variations in the chemical composition of the interstitial and overlying water with regard to the lithology of sediments on the example of lacustrine environment of Lake Wigry. Lake requires special attention because of its complexity. Individual parts of this lake are strongly differentiated in terms of shape, depth and bottom sculptures and water supply, which is associated with their origins. There are also different sedimentation conditions and various external factors that may have a significant impact on the final formation of sediments, on their chemistry and chemical composition of pore solutions occurring in them. All of these make the described lake a unique and universal at the same time, in relation to other lakes of similar origin. The results can also show pore water as a very sensitive indicator pointing to pollutants degree and their transport, which could become a contribution to their use in the monitoring research.

Research area

The study was conducted in north-eastern Poland in the area of Lake Wigry located in Wigry National Park, on the border of the Eastern Suwałki Lake District and the Augustów Plain (Kondracki 1988). It is one of the largest and deepest lakes in Poland (Jańczak 1999). It consists of several parts that differ in size, depth, number of islands, chemical composition of water and the nature of shores. There are both extensive cavities, as well as, irregular or elongated ridges (40–50 m) there. The Czarna Hańcza and Wiatrołūza rivers determine the magnitude as well as dynamic of water flow into Lake Wigry. These rivers deliver an average of 91.7 hm³ of water to the lake per year. Taking into consideration annual cycle of circulation, in the whole Lake Wigry horizontal exchange of water plays a key role. In the period of 2001–2008, Lake Wigry contained 117.829 hm³ of water. River water flowing into the lake constituted 84 % of all inflows and 92 % of all outflows. The ratio between water flowing into the lake and water leaving the lake was 0.92, and difference between sum of water inflow and outflow, called retention, was 0.29 hm m³ (Bajkiewicz–Grabowska 2009). Both the river water and the groundwater that supply the lake are rich in dissolved calcium carbonate, which comes from the dilution of Pleistocene deposits. Wigry is a dimictic lake with summer and winter stagnation and spring and autumn mixion. In the summer, epilimnion reaches to a depth of 8 m and metalimnion to about 12 m. From the depth below a dozen or so meters (hypolimnion), water temperatures are fixed and do not exceed 5–6 °C. The oxygen conditions of water are variable. During the summer stagnation period, the conditions are rather bad, and even anoxic (Rutkowski

et al. 2007). The natural lake resistance to degradation is high due to the reservoir morphometric parameters: large surface area and volume, favourable location of the inflow in relation to the outflow and little influence on lake the nearest area of catchment. The physicochemical processes in the Czarna Hańcza River (dilution, precipitation, sedimentation of mineral and organic compounds) determine the lake water quality (Zdanowski and Hutorowicz 1997).

Both Lake Wigry and other smaller basins in the immediate vicinity of that lake have the same origin. Their present surface was mainly formed by the action of accumulation and deforming during the advance and then deglaciation of Vistulian ice sheet. Lake Wigry in the present form was formed in the Late Pleistocene due to the melting of the dead ice blocks which filled its particular basins called regionally 'płosa' (Ber 2009). Lake Wigry is an unusual, mesotrophic lake, which consists of a few basins connected by straights which together with bays and islands divide water masses into several water regions. Each region of the lake has its own unique water depth. The shape of the lake and the lake bottom is conducive to diverse habitat conditions, in particular in trophy, thermal and oxygen conditions, and water exchange (Kamiński 1999; Rutkowski et al. 2009). Research of carbon dioxide concentration showed that Lake Wigry is not a sink but is a source for the atmospheric CO₂. However, an additional or significant source of CO₂ to lake water can also be sediment (Paprocka and Leśniak 2008).

Sediments of Lake Wigry show a high variability and have been the object of scientific interest dating back to the beginning of the last century (e.g. Stangenberg 1938). These are mostly carbonate–organic formations (calcareous gyttjas), specific for deep areas known as the profundal zone. Pure carbonate sediments (lacustrine chalk) predominate in the littoral zone, while clastic sediment can be found close to the banks. Specific organic-reach sediment occurs locally in the Hańczańska Bay and in dystrophic lakes regionally called 'suchar'. These small brown water lakes are closed systems and are surrounded by forests (Rutkowski et al. 2002, 2003, 2009). For that type of lakes, very typical is sediment called 'dy', which is gelatinous and acidic sediment formed in anaerobic conditions by flocculation and precipitation from dissolved large and insoluble particles of humus materials. Its thickness reaches few meters or more (Huggett 2007; Drzymulska et al. 2014). To assess determination of dystrophy level in 'suchar' polyhumic lakes in Wigry National Park, new limnological index—Hydrochemical Dystrophy Index, *HDI*—was used (Górniak 2004), which is calculated from pH, EC, DIC and DOC in surface water. When *HDI* values are higher than 50, dystrophic conditions occur.

Methods

To assess the variations in the chemical composition of interstitial and overlying water (near the sediment–water interface), seven sampling positions were selected (Fig. 1), taking into account different zones (littoral, profundal and dystrophic) and lithological type of deposits (Table 1). A specific sampling point (W–5) was located in the mixing zone of the water (contaminated by the Czarna Hańcza River with relatively clear water of Lake Wigry). The point W–3 was used for the assessment of hydrogeochemical conditions occurring in dystrophic lakes, which occur in the vicinity of Lake Wigry in relatively large amounts.

Samples of sediment and overlying water were collected from individual points in the area of Lake Wigry. Samples of bottom sediment were taken by a gravity sampler, which allows cores of different lengths (from 30 to 100 cm) to be taken depending on local conditions (Figs. 2, 3, 4, 5). Locations were determined by GPS and the water depth by using an echosounder (FCV, Furuno). Sediment cores were sliced into segments (5, 10, 15 or 20 cm) according to the lithology. Three to four sediment samples were taken from each position. In total, 25 sediment samples and 7 water samples were collected. Interstitial water was isolated by the centrifugation method, using a laboratory centrifuge (*Beckman*) with a rotor placed in the horizontal plane. Centrifugation was performed at 3000 rpm for 20 min.

The analyses of water chemical composition were conducted at the Hydrogeochemical Laboratory in the Department of Hydrogeology and Engineering Geology at Faculty of Geology, Geophysics and Environmental Protection, AGH University of Science and Technology in Kraków. They were performed according to the standard certified analytical quality control procedure. Reagent blanks and certified reference materials were used to control the analytical accuracy. Results from the ICP–MS method were confirmed with F–AAS measurements, which attested to the very good analytical quality of all analysed elements.

The water samples' pH, cations (Na⁺, K⁺, Ca²⁺, Mg²⁺, Fe²⁺, Mn²⁺, Zn²⁺, Co²⁺, Pb²⁺, Cd²⁺, Cr³⁺) and anions (Cl[−], SO₄^{2−}, HCO₃^{2−}, PO₄^{3−}, NO₃[−]) were indicated according to the methodology given in Table 2. The hydrogeochemical type of water was also determined (Figs. 2, 3, 4, 5) in accordance with the Szczukariew–Prikłonski classification (Alekin 1970).

Results and discussion

Lithological types of sediments

The first type of samples constitutes carbonate sediment—lacustrine chalk. It was identified in Płoso Bryzłowskie

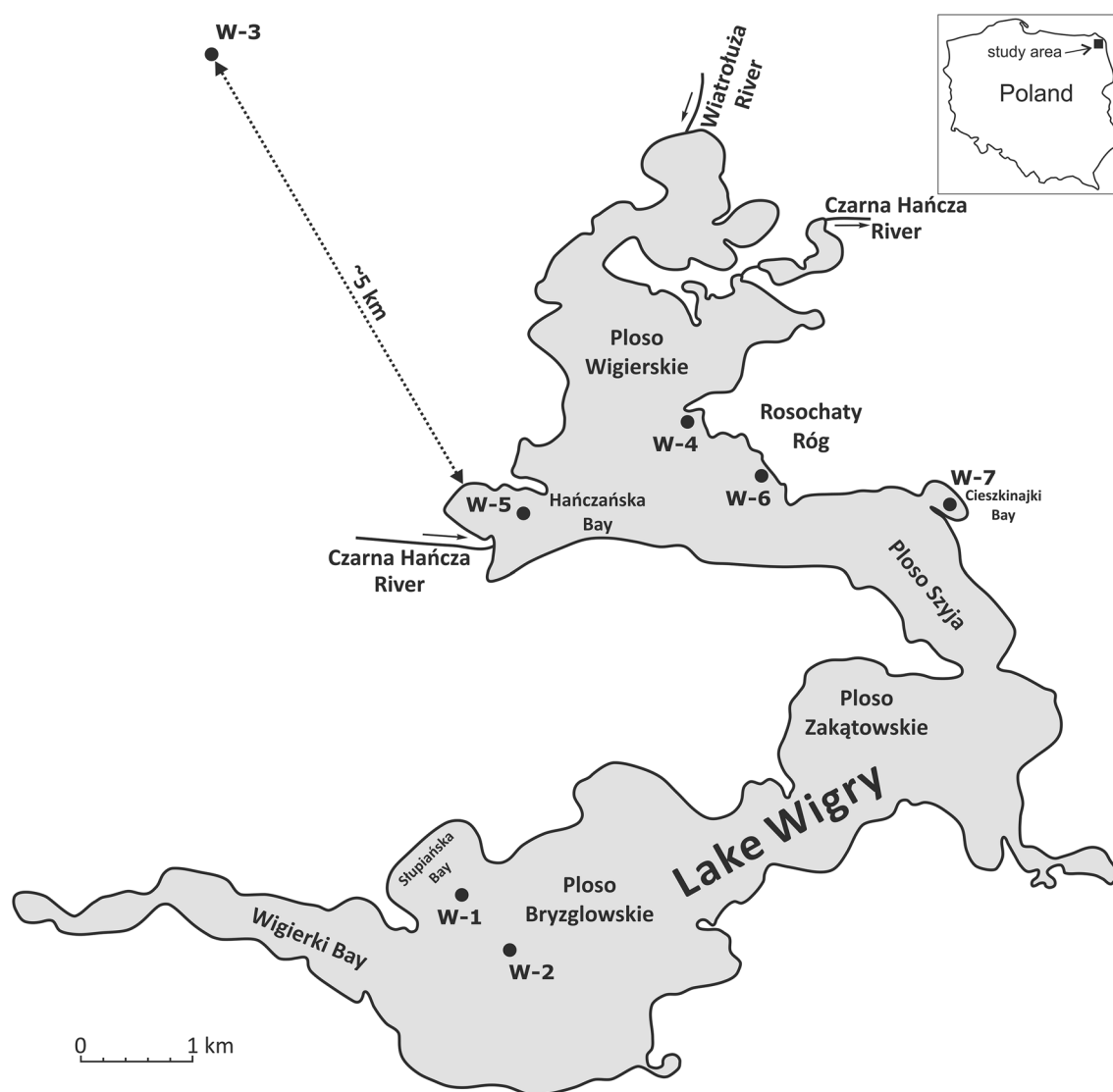


Fig. 1 Locations of sediments and overlying water sampling points

Table 1 Characterization of sediments and overlying water sampling points

Type of sediment	Sampling point	Location*
Lacustrine chalk	W-2	Ploso Bryzglowskie
	W-6	Rosochaty Róg
Calcareous gyttja	W-1	Słupiańska Bay
	W-4	bay in Rosochaty Róg
	W-7	Cieszkinajki Bay
Detritus gyttja	W-5	Hańczańska Bay
Dy	W-3	Suchar—dystrophic lake

* According to Fig. 1

(W-2) and Rosochaty Róg (W-6) (Fig. 1) and had the typical features as described by Rutkowski et al. (2009). It had a bright colour, large contents of clay and silt grain

size-fraction which results in poor permeability for water, a relatively high bulk density ($1.4\text{--}1.5\text{ g/cm}^3$) and the high content ($>90\%$) of calcium carbonate in the form of limestone dust and irregular lumps. Apart from the calcium carbonate, the main component of this sediment was organic substance and quartz.

In the analysed bays, the gyttja samples were found. In Słupiańska Bay (W-1), bay in Rosochaty Róg (W-4) and Cieszkinajki Bay (W-7) (Fig. 1), it was calcareous gyttja. The main component of samples is typical for such sediment in Lake Wigry (Rutkowski et al. 2009); it was also calcium carbonate, but the amount ($54\text{--}87\%$) was less than in lacustrine chalk and organic matter (about 10%). They were very fine grained, resulting in their low permeability. Their water content was very high ($75.6\text{--}95.6\%$) and significantly higher than in the lacustrine chalk; however,

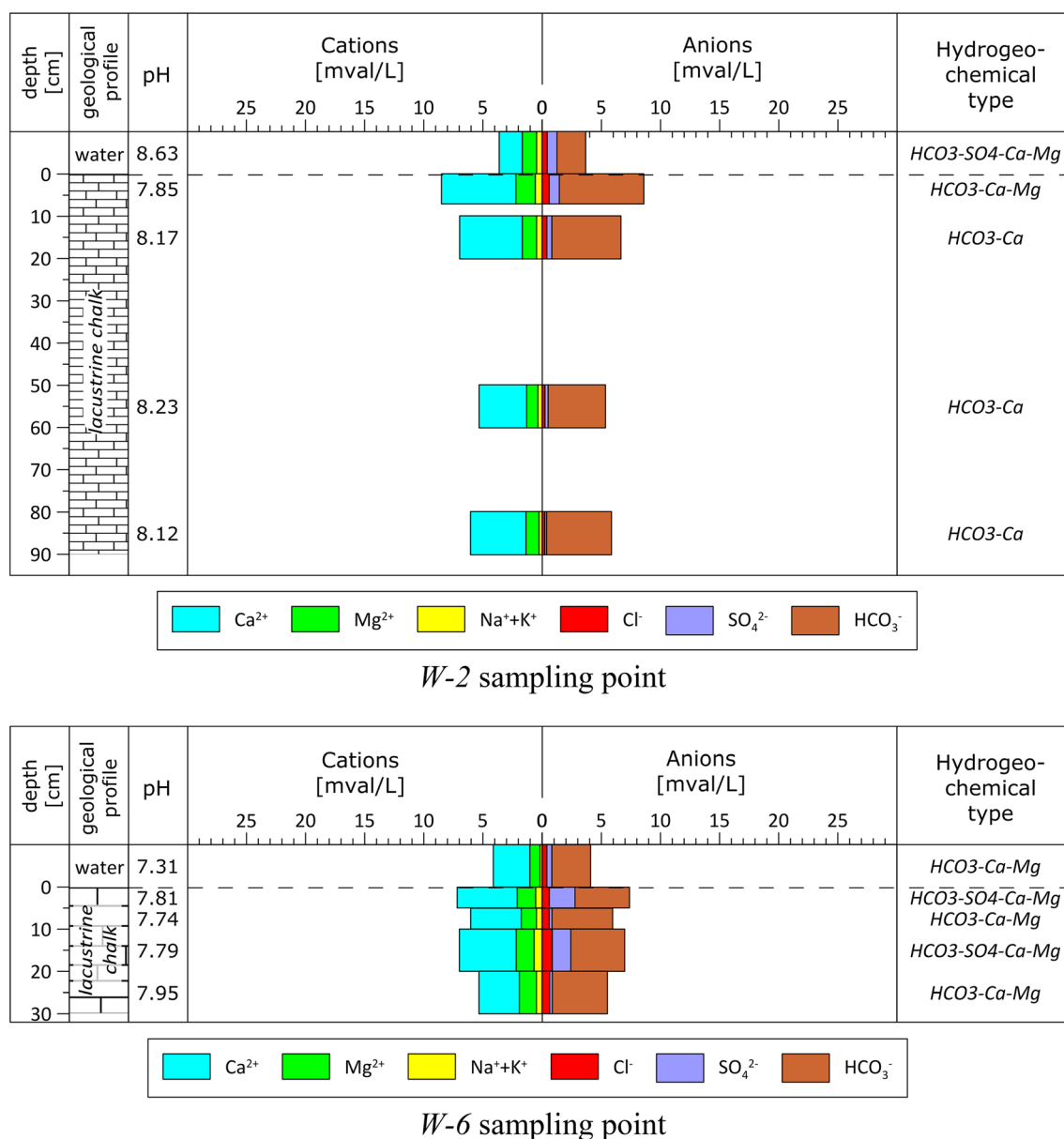


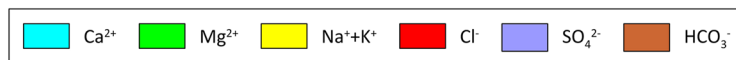
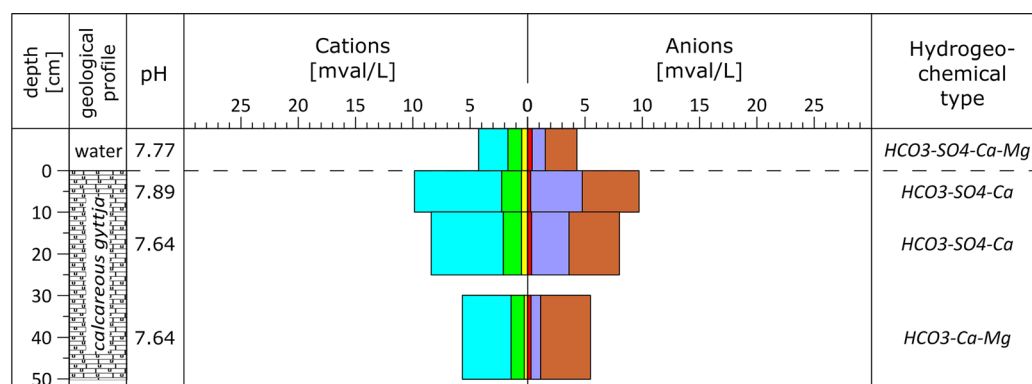
Fig. 2 Variation of chemical composition of the overlying water and interstitial water in the vertical profile of lacustrine chalk type of sediments from Lake Wigry

in contrast, their bulk density was very low: 1.05–1.24 g/cm³. In water separated from these deposits, lower concentrations of calcium and magnesium ions were indicated and connected therewith even slightly lower pH. Additionally, in the profundal zone a high iron and magnesium contents can be seen, which could indicate an enrichment of these metals that are abundant in groundwater, or about the diagenetic processes taking place in the sediments (Salomons and Förstner 1984).

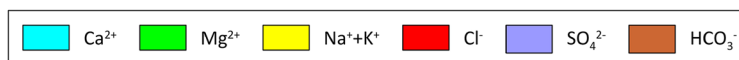
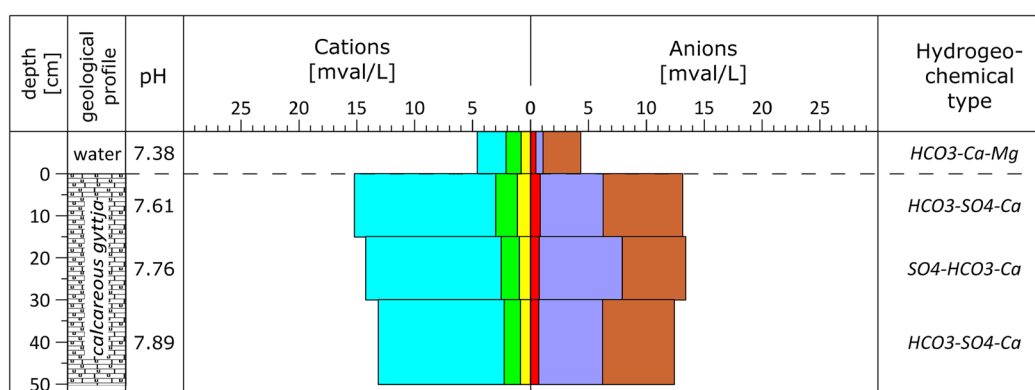
Lake sediment formed directly at the mouth of the Czarna Hańcza River (Hańczańska Bay, W-5, Fig. 1) was described by Rutkowski et al. (2009) as detritus gyttja. It had specific character: it was built from the remains of

plants and has high water content (84.4–91.4 %), low bulk density (1.01–1.05 g/cm³) and low (3.0–13.4 %) CaCO₃ concentration. In these deposits, the influences of pollutants carried with the river were indicated (Aleksander-Kwaterczak and Proszowicz 2007).

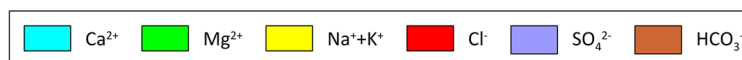
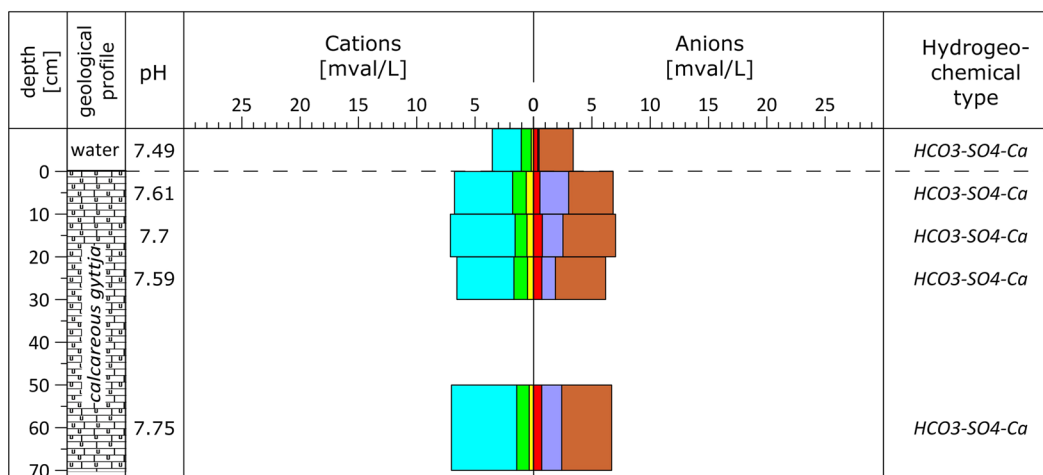
‘Suchar’ is a small dystrophic lake, isolated and without outflow, formed by the melting of blocks of ice during the last ice age (Rutkowski et al. 2009). Sediment samples known as dy were brown in colour and heavily hydrated in surface layers. These were pure organic deposits built with humic acids resulting from the decomposition of organic material and with the absence or a slight admixture of calcium carbonate. Lake with W-3 point (Fig. 1), which



W-1 sampling point



W-4 sampling point



W-7 sampling point

Fig. 3 Variation of chemical composition of the overlying water and interstitial water in the vertical profile of calcareous gyttja type of sediments from Lake Wigry

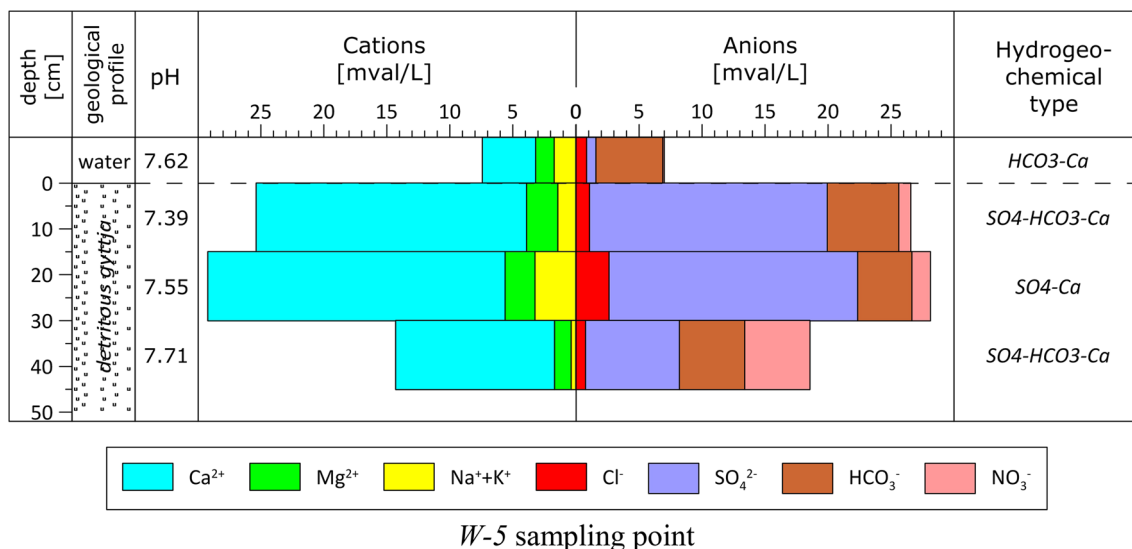


Fig. 4 Variation of chemical composition of the overlying water and interstitial water in vertical profile of detritus gyttja type of sediments from Lake Wigry

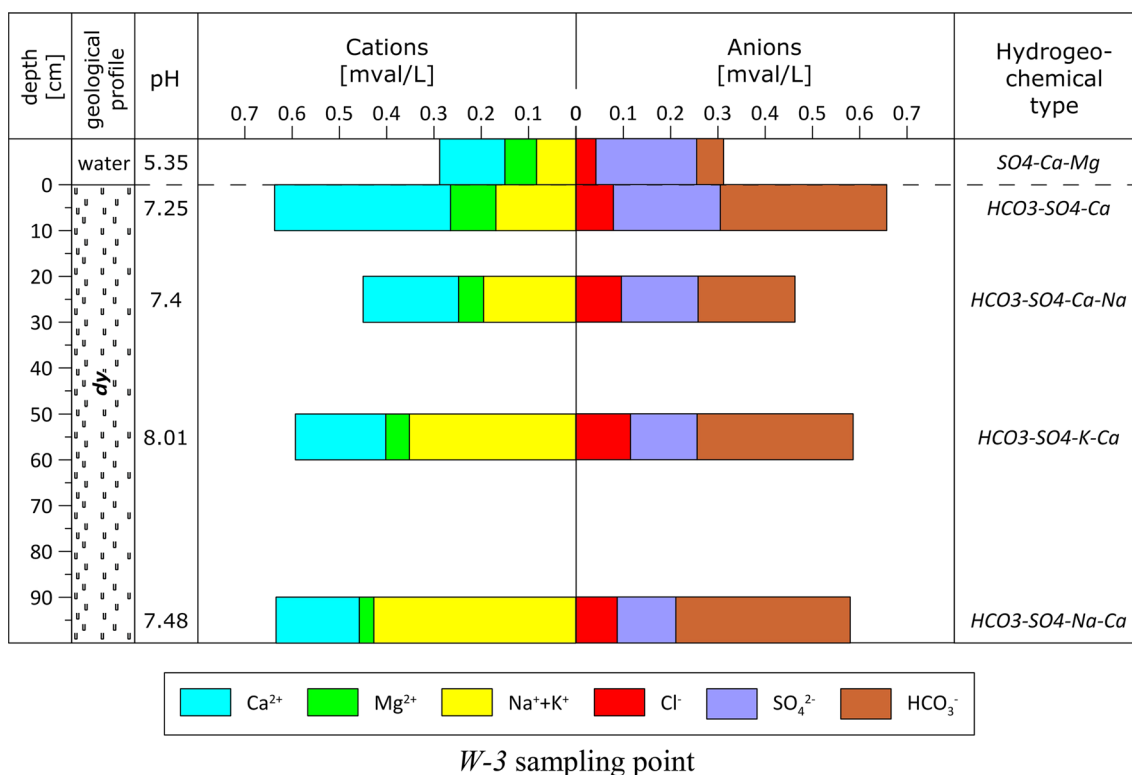


Fig. 5 Variation of chemical composition of the overlying water and interstitial water in the vertical profile of dy type of sediments from dystrophic lake ('suchar')

Table 2 Methods of physicochemical parameters determinations

Parameter	Method	
pH	Potentiometric	PN-C-04540-01:1990
Ions concentration	Inductively Coupled Plasma—Mass Spectrometry (ICP-MS)	PN-EN ISO 17294-2:2006;
		PN-EN ISO 17294-1:2007
Chlorides	Titration (Mohr method)	PN-EN ISO 9297:1994
Nitrates	Spectrophotometric	PN-C-04576-08:1982
Alkalinity	Titration	PN-EN ISO 9963-1:2001+Ap1:2004

according to Górniak (2004) is 'Suchar III', had the most dystrophic conditions ($HDI = 121$).

Hydrochemical characteristic of overlying water

Values of the main physical and chemical parameters (Tables 3, 4) were determined using basic statistical characteristics. There are many methods of statistical analysis for environmental data with low, below the limit of detection, values. The best-known statistical techniques were compared by Antweiler and Taylor (2008), assuming that the data sets have less than 70 % censored data. In the cases at high degrees of censoring, this is greater than 70 % censored data; no technique provided good estimates of summary statistics.

In the case of results presented in the paper, according to Commission Directive 2009/90/EC, during the calculations of mean values, for metals (Table 4) all data below the detection limits (DL) were set to half of these limits values ($1/2$ DL). The data used are archival, and there is not a possibility to present them in the uncensored form. At the same time, the applied method of proceeding does not play a major role, as detection limits adopted are, in general, low and do not affect the overall water quality classification.

Overlying water samples taken from zones with lacustrine chalk type of sediment were characterized by slightly alkaline pH (8.63 in W-2 and 7.31 in W-6; Table 3; Fig. 2). They had a similar hydrogeochemical type: HCO_3 -Ca-Mg in Rosochaty Róg (W-6) and HCO_3 - SO_4 -Ca-Mg in Płoso Bryzglowskie (W-2) and low values of mineralization (270–314 mg/L). There was also very low metals concentration in them (Table 4).

Chemical composition of the overlying water taken from zones above the calcareous gyttja sediments (W-1, W-4 and W-7; Table 3; Fig. 3) was similar to that from above lacustrine chalk. These were multi-ion water of HCO_3 - SO_4 -Ca-Mg, HCO_3 -Ca-Mg or HCO_3 - SO_4 -Ca type. They had a low mineralization (268–334 mg/L) and were weakly alkaline (pH from 7.38 to 7.77). Generally, the slightly higher metal concentrations were found compares with water collected from lacustrine chalk zone (Table 4).

The situation was different with the composition of the water taken from the zone of detritus gyttja deposits (Table 3). Overlying water collected from Hańczańska Bay (W-5, Fig. 4) had the highest mineralization 552 mg/L, HCO_3 -Ca type and low metal concentrations, while water from the dystrophic lake (W-3, Fig. 5) of SO_4 -Ca-Mg type was low mineralized (28 mg/L) and acidic (pH = 5.35). Natural acidification is caused by accumulation of the humus substances and the absence of inflows. There are very good conditions to higher metal concentration in the mobile fractions, especially in the oxides and hydroxides of iron or the organic matter fraction (Kłaviņš et al. 1995; Cudowski 2015) and their migration into overlying water.

Hydrogeochemical characterization of interstitial water in sediment cores

Hydrogeochemical background values were used for the physicochemical composition assessment of interstitial water. It is a range of concentrations of the analysed substance or a range of hydrochemical features values, which is characteristic for a particular environment and is limited by the lower and upper limits of concentration beyond which are anomalous values (Dowgiałło et al. 2002). The limit values of hydrogeochemical background (Table 5) were determined as a probability of 16 % percentile as lower limit and 84 % percentile as upper limit (according to Szczepańska and Kmiecik 2005). This procedure is the most often used in Poland and allows the comparison of own results with the results obtained for other locations.

The values of the hydrogeochemical background were determined also for selected metal ions (Fe^{2+} , Mn^{2+} , Zn^{2+} , Co^{2+} , Pb^{2+} , Cd^{2+} , Cr^{3+}), using the statistical method (Table 6). As described above (according 2009/90/EC) during the calculations for mean values of metal, data below detection limit (DL) were changed to half value of limit ($1/2$ DL).

Differences in physical and chemical composition were found between interstitial and overlying water in every individual position. Generally, some differences were found between the water chemical composition of the

Table 3 Statistical characteristic of the main physical and chemical parameters of overlying water

Parameter	Type of sediment*	Min	Max	Mean value
pH	Lacustrine chalk	7.31	8.63	–
	Calcareous gyttja	7.38	7.77	–
	Detritus gyttja	7.62	7.62	–
	Dy	5.35	5.35	–
Mineralization [mg/L]	Lacustrine chalk	270.6	314.3	292.5
	Calcareous gyttja	268.1	334.0	309.6
	Detritus gyttja	552.3	552.3	552.3
	Dy	28.0	28.0	28.0
Ca ²⁺ [mg/L]	Lacustrine chalk	39.16	61.83	50.50
	Calcareous gyttja	49.60	51.37	50.25
	Detritus gyttja	84.76	84.76	84.76
	Dy	2.76	2.76	2.76
Mg ²⁺ [mg/L]	Lacustrine chalk	10.38	14.80	12.59
	Calcareous gyttja	10.44	16.05	13.68
	Detritus gyttja	17.89	17.89	17.89
	Dy	0.81	0.81	0.81
Na ⁺ [mg/L]	Lacustrine chalk	3.15	8.80	5.98
	Calcareous gyttja	3.19	16.05	9.30
	Detritus gyttja	33.81	33.81	33.81
	Dy	0.50	0.50	0.50
K ⁺ [mg/L]	Lacustrine chalk	1.38	2.76	2.07
	Calcareous gyttja	1.67	4.87	3.38
	Detritus gyttja	9.87	9.87	9.87
	Dy	2.37	2.37	2.37
Cl [−] [mg/L]	Lacustrine chalk	14.0	15.1	14.6
	Calcareous gyttja	13.2	16.4	14.5
	Detritus gyttja	29.9	29.9	29.9
	Dy	1.50	1.50	1.50
SO ₄ ^{2−} [mg/L]	Lacustrine chalk	20.0	39.5	29.8
	Calcareous gyttja	5.0	55.4	29.9
	Detritus gyttja	35.6	35.6	35.6
	Dy	10.2	10.2	10.2
HCO ₃ [−] [mg/L]	Lacustrine chalk	147.6	200.0	173.8
	Calcareous gyttja	167.9	198.0	181.3
	Detritus gyttja	324.0	324.0	324.0
	Dy	3.5	3.5	3.5
PO ₄ ^{3−} [mg/L]	Lacustrine chalk	0.03	0.03	0.03
	Calcareous gyttja	0.05	0.06	0.06
	Detritus gyttja	0.13	0.13	0.13
	Dy	0.12	0.12	0.12

* Sampling points—see Table 1

carbonate and from organic sediments, which was also confirmed in the studies of Woroniecka–Stasiak (1980).

There was just a slight difference in pH values for water samples taken from **lacustrine chalk** zones (W–2 and W–6; Table 5; Fig. 2). Such sediment characteristic was reflected in interstitial water composition. Water had high pH values (from 7.74 to 8.23); however, there was a slight

pH increase as the depth increased, whereas in samples from W–2 position, the pH values variation with depth was irregular.

Interstitial water in samples of **lacustrine chalk** had a low content of calcium and magnesium and a relatively low mineralization. In such environmental conditions, most metals are precipitated and exist in the forms associated

Table 4 Statistical characteristic of selected heavy metals in overlying water

Parameter	Type of sediment*	Min	Max	Mean value
Fe ²⁺ [mg/L]	Lacustrine chalk	<0.010	0.030	0.020
	Calcareous gyttja	0.018	0.021	0.020
	Detritus gyttja	0.177	0.177	0.177
	Dy	0.749	0.749	0.749
Mn ²⁺ [mg/L]	Lacustrine chalk	<0.003	<0.003	<0.003
	Calcareous gyttja	<0.003	0.027	0.010
	Detritus gyttja	0.014	0.014	0.014
	Dy	0.085	0.085	0.085
Zn ²⁺ [mg/L]	Lacustrine chalk	0.0012	0.0024	0.0018
	Calcareous gyttja	0.0016	0.0064	0.0036
	Detritus gyttja	0.011	0.011	0.011
	Dy	0.019	0.019	0.019
Co ²⁺ [mg/L]	Lacustrine chalk	<0.0002	<0.0002	<0.0002
	Calcareous gyttja	<0.0002	<0.0002	<0.0002
	Detritus gyttja	0.0005	0.0005	0.0005
	Dy	<0.0002	<0.0002	<0.0002
Pb ²⁺ [mg/L]	Lacustrine chalk	<0.0001	<0.0001	<0.0001
	Calcareous gyttja	<0.0001	<0.0001	<0.0001
	Detritus gyttja	0.0003	0.0003	0.0003
	Dy	0.0006	0.0006	0.0006
Cd ²⁺ [mg/L]	Lacustrine chalk	<0.0003	<0.0003	<0.0003
	Calcareous gyttja	<0.0003	<0.0003	<0.0003
	Detritus gyttja	<0.0003	<0.0003	<0.0003
	Dy	<0.0003	<0.0003	<0.0003
Cr ³⁺ [mg/L]	Lacustrine chalk	<0.005	<0.005	<0.005
	Calcareous gyttja	<0.005	<0.005	<0.005
	Detritus gyttja	<0.005	<0.005	<0.005
	Dy	<0.005	<0.005	<0.005

* Sampling points—see Table 1

with the solid phase; hence, their concentrations in water are very low. Hydrogeochemical types of water were similar—predominant ions are Ca²⁺, SO₄²⁻ and HCO₃⁻; hence, the most common type of water was HCO₃-Ca, HCO₃-Ca-Mg or HCO₃-SO₄-Ca-Mg. Concentrations of analysed metals in water were very low, lower if compared with other analysed type of sediments (Table 6), which can be connected with alkaline environment (Salomons and Frstner 1984; Elder 1989; Salomons 1995; Wällstedt and Borg 2003) and low metals concentration in carbonate sediments of Lake Wigry (Aleksander-Kwaterczak et al. 2009; Prosoiwicz et al. 2009; Aleksander-Kwaterczak and Kostka 2011).

The pH of the interstitial water collected from **calcareous gyttja** samples (Table 5; Fig. 3) was very similar and changed in the range from 7.59 to 7.89. There was also just a slight increase in the pH values in relation to overlying water and a general increase of pH with the increasing sediment depth. A large variation of

mineralization was determined: from lower values founded for the solutions from W-1 positions—479 mg/L in the depth from 30 to 50 cm, to 1119 mg/L in the depth from surface to 15 cm in the W-4 position. The mineralization differed slightly in the W-7 position (from 512 to 580 mg/L). In the position of W-1, mineralization decreased with depth from 762 mg/L to 479 mg/L. Interstitial water samples were more homogeneous in terms of their chemical type; the dominant type was HCO₃-SO₄-Ca and only in W-1 position in the depth from 30 to 50 cm HCO₃-Ca-Mg type of water occurred.

Samples collected from **calcareous gyttja** had the highest concentrations of Fe²⁺ ions (mean value 0.156 mg/L, Table 6). Samples from calcareous gyttja statistically hold the highest mean concentrations of Mn²⁺ (0.162 mg/L), Zn²⁺ (0.04 mg/L), Co²⁺ (0.0004 mg/L), Pb²⁺ (0.0026 mg/L), Cr³⁺ (0.008 mg/L) and Cd²⁺ (0.00033 mg/L).

The chemical composition of interstitial water collected from **detritus gyttja** in Hańczańska Bay varied as

Table 5 Hydrogeochemical background values of the main physical and chemical parameters of interstitial water

Parameter	Type of sediment*	Min	Max	Interval	Mean value	Percentile 16 %	Percentile 84 %
pH	Lacustrine chalk	7.74	8.23	0.49	–	7.79	8.16
	Calcareous gyttja	7.59	7.89	0.30	–	7.61	7.83
	Detritus gyttja	7.39	7.71	0.32	–	7.44	7.66
	Dy	7.25	8.01	0.76	–	7.32	7.76
Mineralization [mg/L]	Lacustrine chalk	445.6	711.6	266.0	544.5	481.3	586.7
	Calcareous gyttja	479.0	1118.7	639.7	717.7	522.6	997.0
	Detritus gyttja	1102.4	2031.7	929.3	1670.9	1350.8	1982.7
	Dy	49.5	69.1	19.6	61.4	55.2	67.4
Ca ²⁺ [mg/L]	Lacustrine chalk	68.4	126.0	57.7	94.9	81.3	105.8
	Calcareous gyttja	85.4	245.0	159.6	148.3	98.9	227.2
	Detritus gyttja	252.9	472.7	219.8	385.3	309.7	459.1
	Dy	3.5	7.5	3.9	4.7	3.7	5.8
Mg ²⁺ [mg/L]	Lacustrine chalk	11.5	20.0	8.6	16.3	13.5	18.9
	Calcareous gyttja	12.3	22.8	10.5	16.7	13.3	20.3
	Detritus gyttja	16.1	30.2	14.2	25.1	20.2	29.8
	Dy	0.4	1.2	0.8	0.7	0.5	0.9
Na ⁺ [mg/L]	Lacustrine chalk	3.8	14.1	10.4	8.8	5.3	10.7
	Calcareous gyttja	5.1	22.8	17.7	11.6	6.6	18.4
	Detritus gyttja	7.1	28.9	21.8	20.7	13.2	28.0
	Dy	1.9	8.4	6.4	3.7	1.9	5.6
K ⁺ [mg/L]	Lacustrine chalk	2.1	5.3	3.1	3.5	2.3	5.0
	Calcareous gyttja	2.4	6.3	3.9	4.7	3.9	5.8
	Detritus gyttja	2.3	77.8	75.5	30.4	5.1	56.5
	Dy	2.5	10.5	8.0	4.9	2.8	7.1
Cl [−] [mg/L]	Lacustrine chalk	7.6	30.3	22.7	18.1	9.0	21.9
	Calcareous gyttja	9.5	29.8	20.3	20.9	11.0	26.6
	Detritus gyttja	26.8	93.1	66.3	52.7	30.4	75.5
	Dy	2.8	4.1	1.3	3.4	2.9	3.8
SO ₄ ^{2−} [mg/L]	Lacustrine chalk	7.3	104.0	96.7	35.4	11.2	71.0
	Calcareous gyttja	40.5	344.2	303.8	162.2	67.1	262.4
	Detritus gyttja	357.8	947.2	589.4	737.0	533.2	934.0
	Dy	6.00	10.8	4.9	7.8	6.4	9.4
HCO ₃ [−] [mg/L]	Lacustrine chalk	278.0	436.2	158.2	322.4	281.4	353.9
	Calcareous gyttja	233.0	419.0	186.0	299.8	261.9	358.6
	Detritus gyttja	263.0	346.0	83.0	308.3	280.0	336.4
	Dy	12.5	22.5	10.0	19.2	16.1	22.0
PO ₄ ^{3−} [mg/L]	Lacustrine chalk	0.06	0.08	0.02	0.07	0.06	0.08
	Calcareous gyttja	0.10	0.42	0.32	0.23	0.15	0.34
	Detritus gyttja	0.35	0.57	0.22	0.47	0.39	0.54
	Dy	0.21	1.33	1.12	0.61	0.21	1.01

* Sampling points—see Table 1

compared to water from other sediments. It had a much lower pH, which clearly results from the high content of organic matter, and there was a slight pH increase with the depth increasing (Fig. 4). It also had a different chemical composition; dominated ions are SO₄^{2−} or Ca²⁺ and SO₄–

Ca or SO₄–HCO₃–Ca type of water. The highest concentration of Zn²⁺, Co²⁺, Cd²⁺ (Table 6) and a relatively higher mineralization (Table 5) were discovered in them. In the depth range of 15–30 cm, much higher values of mineralization (2032 mg/L) were found, and in the depth

Table 6 Hydrogeochemical background values of selected heavy metals in interstitial water

Parameter	Type of sediment*	Min	Max	Interval	Mean value	Percentile 16 %	Percentile 84 %
Fe ²⁺ [mg/L]	Lacustrine chalk	0.011	0.070	0.059	0.043	0.014	0.070
	Calcareous gyttja	0.026	0.860	0.834	0.156	0.033	0.193
	Detritus gyttja	0.032	0.046	0.014	0.039	0.034	0.044
	Dy	0.072	0.290	0.218	0.150	0.081	0.220
Mn ²⁺ [mg/L]	Lacustrine chalk	0.003	0.036	0.033	0.017	0.004	0.033
	Calcareous gyttja	0.013	0.462	0.449	0.162	0.020	0.355
	Detritus gyttja	0.124	0.242	0.118	0.173	0.133	0.213
	Dy	0.023	0.061	0.038	0.036	0.025	0.048
Zn ²⁺ [mg/L]	Lacustrine chalk	0.002	0.021	0.019	0.013	0.004	0.017
	Calcareous gyttja	0.018	0.053	0.035	0.040	0.029	0.052
	Detritus gyttja	0.097	0.147	0.050	0.124	0.107	0.141
	Dy	0.040	0.120	0.080	0.075	0.043	0.108
Co ²⁺ [mg/L]	Lacustrine chalk	0.0002	0.0007	0.0005	0.0003	0.0002	0.0005
	Calcareous gyttja	0.0002	0.0005	0.0003	0.0004	0.0003	0.0005
	Detritus gyttja	0.0006	0.0009	0.0003	0.0008	0.0007	0.0009
	Dy	0.0002	0.0002	0.0000	0.0002	0.0002	0.0002
Pb ²⁺ [mg/L]	Lacustrine chalk	<0.0001	0.0022	–	0.0008	–	–
	Calcareous gyttja	0.0001	0.0105	0.0103	0.0026	0.0002	0.0068
	Detritus gyttja	0.0001	0.0002	0.0001	0.0001	0.0001	0.0002
	Dy	0.0092	0.0276	0.0184	0.0174	0.0118	0.0231
Cd ²⁺ [mg/L]	Lacustrine chalk	<0.0003	0.0005	–	0.0002	–	–
	Calcareous gyttja	<0.0003	0.0008	–	0.0003	–	–
	Detritus gyttja	0.0006	0.0007	0.0001	0.0007	0.0006	0.0007
	Dy	<0.0003	<0.0003	–	<0.0003	–	–
Cr ³⁺ [mg/L]	Lacustrine chalk	<0.005	<0.005	–	<0.005	–	–
	Calcareous gyttja	<0.005	0.040	–	0.0078	–	–
	Detritus gyttja	<0.005	0.0071	–	0.0045	–	–
	Dy	<0.005	<0.005	–	<0.005	–	–

* Sampling points—see Table 1

range of 30–45 cm higher concentration of nitrates (103 mg/L) were discovered. Such situation is caused by sewage treatment plant, localized near the Czarna Hańcza River, which inflow to Lake Wigry close to W-5 point. This is specific for environment impacted by human land use, first of all industrial wastewater, domestic sewage discharge and runoff from agricultural soil (e.g. Kerfoot et al. 1994; Keller et al. 2007).

This water, particularly in the upper part of the sediment, may have a tendency towards seasonal variation. Their enrichment in phosphorus and nitrogen depends on the deposit in *Hańczańska Bay* brought by the Czarna Hańcza River (Zdanowski and Hutorowicz 1997). The study of the chemical composition of water from this river (Migaszewski et al. 2003) showed an oversize in the ammonium and nitrate content, presumably associated with a close proximity to rural households and tributary water

from sewage treatment plants in nearest located town Suwałki. This influences the fact that eutrophication processes are the greatest threat to the quality of water and sediment in Lake Wigry. Chemical composition of both surface water and interstitial water is greatly influenced by the organic matter content in the sediment (Thresh et al. 1944). Sulphate is generally present in low concentrations in lakes. However, within eutrophic lakes where organic matter availability is higher, sulphate concentrations increase (Holmer and Storkholm 2001). General decrease of SO₄²⁻ ion concentration in interstitial solution with sediment depth can be explained by sulphate reduction. Furthermore, during remineralization of organic matter, microbes in the sediment take up (Carman and Rahm 1997).

Interstitial water samples from isolated **dystrophic lake** (W-3; Table 5, Fig. 5) had definitely different chemical

compositions. Their mineralization was very low (49–69 mg/L), and pH changed from 7.25 to 8.01. Chemical composition was dominated by ions: HCO_3^- , SO_4^{2-} , Ca^{2+} and Na^+ , but their hydrogeochemical type changed with depth, from the $\text{HCO}_3\text{--Ca--SO}_4$, via $\text{HCO}_3\text{--SO}_4\text{--Ca--Na--HCO}_3$ and $\text{SO}_4\text{--K--Ca}$, to the $\text{HCO}_3\text{--SO}_4\text{--Na--Ca}$.

In interstitial water from sediment of dystrophic lake, high concentrations of Fe^{2+} (0.15 mg/L) and the highest mean concentrations of Pb^{2+} (0.0174 mg/L) occurred (Table 6). Although overlying water was acidic and had very low mineralization, it was determined that interstitial water pH increased and water composition strongly changed with the depth. This can be due to lithological variability of sediment, and decrease content of organic matter with depth and increase content of calcium carbonate.

Variation of selected metal concentrations in water samples in the individual sediment segment within the vertical profile and overlying water is shown on Fig. 6. The forms of iron and manganese are considered to play a major role in the availability of nutrients and trace metals in interstitial water. Under aerobic conditions, they are found as Fe^{3+} and Mn^{4+} , respectively, and as insoluble oxide/hydroxides, which have very large surface areas. Many trace elements and oxy-anions can be adsorbed in such surfaces. When oxides undergo reduction (to Fe^{2+} and Mn^{2+}), a large portion of trace elements undergo

dissolution to be released into the pore water (Santschi 1990). Gu et al. (1996) showed that natural organic matter plays a role in stabilizing iron oxide colloids. Lovley et al. (1996) showed that humic acids can indirectly affect the distribution of iron in sediments by stimulating microbial reduction of Fe^{3+} . In our studies, the highest concentration of Fe^{2+} ions occurred in overlying water in W-5 (*Hańczańska Bay*) and in W-3 (dystrophic lake) positions. Also in these locations, the highest organic matter content was stated. The concentration of iron in the interstitial water generally decreased with depth. An exception was found in water collected from the sediment in W-1 position, where the metal concentration increased with depth. In turn, the concentration of Mn^{2+} ions decreased with depth or to a certain depth increased and then further decreased (Fig. 6). For the ions of other metals, it was generally observed that concentrations decreased with the depth of sampling. The relatively higher concentrations were noted for samples collected from a depth of 40–50 cm. The exceptions were samples from dystrophic lake, in which the concentration variability in function of depth is characterized by distinct irregularity.

In the region of Lake Wigry, some interstitial water analyses separated from the sands were investigated by Stangenberg (1934) and Rutkowski et al. (2006). The high permeability rate of such deposits and the easy exchange of

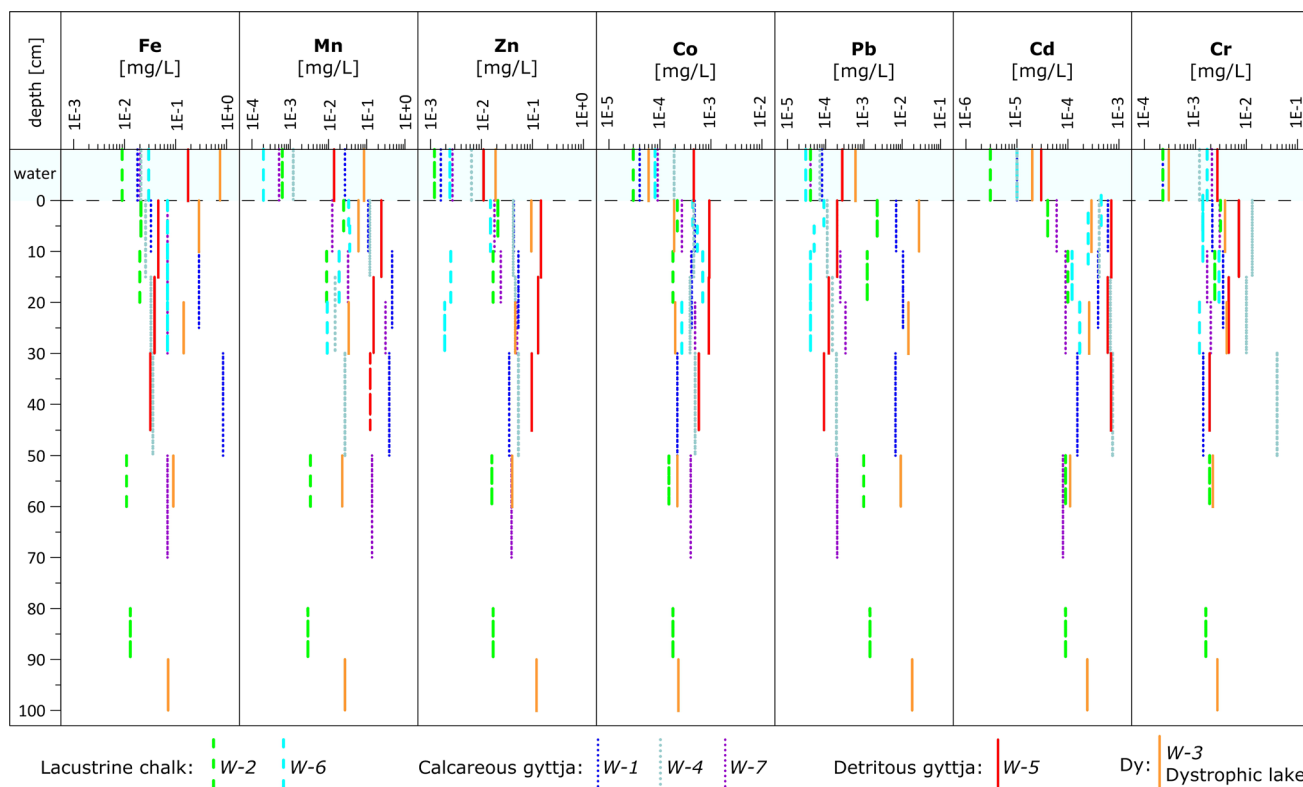


Fig. 6 Variation of selected metal concentration in overlying water and interstitial water in the vertical profile of sediments from Lake Wigry

water suggest that this is almost the same chemical composition of interstitial and overlying water.

It should be also taken into account the fact that first of all in the littoral zone the changes in the distribution of trace metals between the solid particles and interstitial water, and at the border of water and sediments may be also affected by bioturbation (Vale and Sundby 1998; Ciutat et al. 2005).

Conclusions

Studies of interstitial solutions collected from different zones and lithological type of sediments showed marked chemical composition differences between the carbonate and organic sediments. The highest mineralization (2032 mg/L) was generally found in the interstitial water obtained from the detritus gyttja localized in the shallow positions, which are more exposed to pollution. The highest trace metals and nutrients content in the sediments and interstitial water found in the Czarna Hańcza River estuary were a result of the input from increasing industrial wastewater, domestic sewage discharge and runoff from agricultural soil.

A greater resistance to adsorption of contaminants characterizes sediments composed of the lacustrine chalk; mineralization of their pore solutions was significantly lower. This fact is probably also affected by very low mineralization and chemical composition of overlying water and sediment.

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